



DECLARATION

I, Ryu MIYAMOTO, c/o the Inoue & Associates of 3rd Floor, Akasaka Habitation Building, 3-5, Akasaka 1-chome, Minato-ku, Tokyo, Japan do solemnly and sincerely declare that I am conversant with the Japanese and English languages and that I believe:

that the expression "--trade name: RM-65;--" should be inserted before "manufactured and sold by MITENI S.p.A., Italy" at page 10, line 3 of the English specification;

that the expression " $\times 10,000,000$ " at page 29, line 18 of the English specification should be amended to "-- $\times 100,000$ --;

that the expression " $(4S/\mu)^{0.5}$ " at page 30, line 5 of the English specification should be amended to "-- $(4S/\pi)^{0.5}$ --; and

that the expression "Organic sulfonic acid" at page 64, line 12 of the English specification should be amended to "--Alkali metal salt of an organic sulfonic acid--.

These amendments are merely corrections of inadvertent errors which occurred at the time of the translation into English of the original PCT specification. The attached copies of revised pages 10, 29, 30 and 64 of the English specification are true and correct translations of the corresponding pages of the international patent application No.

PCT/JP02/13404. The English description "trade name: RM-65;" in the English specification at page 10, line 3 is a correct English translation of the Japanese description "商品名:「RM-65」" in the original Japanese PCT specification at page 6, lines 20-21. The English description " $\times 100,000$ " in the English specification at page 29, line 18 is a correct English translation of the Japanese description "10万倍" in the original Japanese PCT specification at page 24, line 6. The English description " $(4S/\pi)^{0.5}$ " in the English specification at page 30, line 5 is a correct English translation of the Japanese description " $(4S/\pi)^{0.5}$ " in the original Japanese PCT specification at page 24, line 18. The English description "Alkali metal salt of an organic sulfonic acid" in the English specification at page 64, lines 12-13 is a correct English translation of the Japanese description "有機スルホン酸アルカリ金属塩" in the original Japanese PCT specification at page 51, line 15.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of

the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

February 9, 2005

(Date)

Ryu Miyamoto

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tion obtained in substantially the same manner as in Example 4, except that component (C) was changed to potassium perfluorobutane sulfonate (trade name: RM-65; manufactured and sold by MITENI S.p.A., Italy), and

5 which was subjected to the 20 mm Vertical Burning Test described in UL-Subject 94. In the 20 mm Vertical Burning Test, the test specimen was self-extinguished 3 seconds after the second contact (10 seconds) with fire, and the flame retardancy of the test specimen was "V-0".

10 In the photomicrograph of Fig. 1, black portions are branched metal oxide particles (B) and the remainder is a matrix comprising an aromatic polycarbonate (A), an alkali metal salt (C) of an organic sulfonic acid and a fluoropolymer (D). Further, on the photomicrograph of  
15 Fig. 1, the total area of the metal oxide particles as component (B) is 56 %, based on the area of the photomicrograph.

Fig. 2 is a photomicrograph (taken using a scanning probe microscope) of the surface of a 1/16 inch-thick test specimen which was produced by molding the  
20 same resin composition as that of Fig. 1, and which was subjected to the 20 mm Vertical Burning Test described in UL-Subject 94. In the 20 mm Vertical Burning Test, the test specimen was self-extinguished 6 seconds after  
25 the second contact (10 seconds) with fire, and the

Next, an explanation is given below with respect to component (B) used in the resin composition of the present invention.

Component (B) is branched metal oxide particles.

5           In the present invention, the "branched metal oxide particles" mean aggregates and/or agglomerates of metal oxide primary particles, in which the metal oxide primary particles are bonded together to form branched chains.

10           In the present invention, the "particles" mean those which are observed as distinct particles in the transmission electron microscopy (TEM) performed with respect to an ultrathin specimen of a resin composition, or in the scanning probe atomic force microscopy (SPM)  
15           performed with respect to a surface or cross-section of a molded article of the resin composition (magnification in each of the microscopies is generally from  $\times 10,000$  to  $\times 100,000$ ). In each of the above-mentioned microscopies, the branched metal oxide particles (B)  
20           which are, as mentioned above, in the form of aggregates and/or agglomerates, are observed as distinct particles.

          In the present invention, the particle diameter of branched metal oxide particles (B) can be measured by  
25           the above-mentioned TEM or SPM as follows. With re-

spect to each of 100 or more particles on a TEM or SPM photomicrograph of the resin composition, the area (S) thereof is measured. Using the measured areas (S) of the particles, the diameter of each of the particles is calculated by the formula  $(4S/\pi)^{0.5}$ . Further, using the above-mentioned photomicrograph, the particle diameter distribution can also be determined.

Examples of branched metal oxide particles (B) include particles of silicon oxide, titanium oxide, aluminum oxide, zinc oxide, cerium oxide, yttrium oxide, zirconium oxide, tin oxide, iron oxide, magnesium oxide, manganese oxide and holmium oxide, which are in the form of aggregates and/or agglomerates of primary particles, in which the primary particles are bonded together to form branched chains.

Of these branched particles, branched particles of silicon oxide, titanium oxide and aluminum oxide are preferred, and branched particles of silicon oxide are most preferred.

In the resin composition of the present invention, for causing the resin composition to exhibit a high flame retardancy and an excellent impact resistance, it is necessary that at least 70 %, preferably 75 % or more, still more preferably 80 % or more, most preferably 90 % or more, of the branched metal oxide particles

(fumed silica which has not been subjected to surface treatment) (trade name: Aerosil 200; manufactured and sold by Nippon Aerosil Co., Ltd., Japan) produced by the dry method.

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(Oxide particles 3): Modified, branched titanium oxide particles (trade name: Aerosil T805; manufactured and sold by Nippon Aerosil Co., Ltd., Japan) obtained by subjecting titanium oxide particles (produced by the dry method) to surface treatment with octylsilane.

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3. Component (C): Alkali metal salt of an organic sulfonic acid

The following substance ( $C_4F_9SO_3K$ ) is used as component (C).

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( $C_4F_9SO_3K$ ): Potassium perfluorobutanesulfonate (trade name: MEGAFACE F-114; manufactured and sold by DAINIPPON INK & CHEMICALS INC., Japan).

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4. Component (D): Fluoropolymer

The following substance (PTFE/AS) is used as component (D).

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(PTFE/AS): A powder mixture (trade name: Blendex 449; manufactured and sold by GE Speciality Chemicals

カーボネート樹脂組成物であって、該金属酸化物粒子（B）が、該成分（A）、（B）及び（C）の混合物中に分散してなり、該金属酸化物粒子（B）の少なくとも70%が、10～200nmの範囲内に粒子径を有する、ことを特徴とする難燃性芳香族ポリカーボネート樹脂組成物が、臭素系難燃剤やリン系難燃剤を用いなくとも優れた難燃性を発揮し、且つ芳香族ポリカーボネートが本来有する耐熱性や耐衝撃性を損ねることなく、優れた熔融安定性を発揮することを見出した。この知見に基づき、本発明を完成した。

従って、本発明の1つの主たる目的は、臭素系難燃剤やリン系難燃剤を用いなくとも優れた難燃性を発揮し、且つ芳香族ポリカーボネートが本来有する耐熱性や耐衝撃性を損ねることなく、優れた熔融安定性を発揮する難燃性芳香族ポリカーボネート樹脂組成物を提供することにある。

本発明の上記及びその他の諸目的、諸特徴ならびに諸利益は、添付の図面を参照しながら行う以下の詳細な説明及び請求の範囲から明らかになる。

#### 図面の簡単な説明

図1は、成分（C）をイタリア国Miteni社製のパーフルオロブタンスルホン酸カリウム塩（商品名：「RM-65」）に変更する以外は実施例4と同様の方法で得られた難燃性芳香族ポリカーボネート樹脂組成物を成形して得られた



(aggregate)、及び／又は集塊粒子(agglomerate)である。

本発明において「粒子」とは、透過型電子顕微鏡(TEM)を用いて樹脂組成物の超薄切片を観察するか、あるいは走査型プローブ原子間力顕微鏡(SPM)を用いて樹脂組成物の成形体表面もしくは切出面を観察した際(TEM及びSPMでの観察は通常1万～10万倍で行う)に、独立した粒子として観察されるものを意味する。本発明で用いる金属酸化物粒子(B)は、上記したように凝集粒子(aggregate)、及び／又は集塊粒子(agglomerate)であるが、上記の方法で観察した際に独立した粒子として観察される。

本発明では、分岐構造を有する金属酸化物粒子(B)の粒子径は、上記した透過型電子顕微鏡(TEM)による観察、あるいは走査型プローブ原子間力顕微鏡(SPM)による観察によって測定することができる。すなわち、上記の顕微鏡観察において写真撮影を行い、得られた顕微鏡写真から樹脂組成物中における100個以上の粒子に対して個々の粒子径を計測する。さらに粒子径分布を求める。粒子径は、粒子の面積Sを求め、Sを用いて、 $(4S/\pi)^{0.5}$ を粒子径とする。

分岐構造を有する金属酸化物粒子(B)として、例えば、金属酸化物の一次粒子が分岐した鎖状に結合してなる凝集粒子あるいは集塊粒子の形態を有する酸化珪素、酸化チタン、酸化アルミニウム、酸化亜鉛、酸化セリウム、酸化イットリ

2. 成分 (B) : 分岐構造を有する金属酸化物粒子

(酸化物粒子 - 1)

乾式法で得られ、ポリジメチルシロキサンで表面処理された分岐構造を有する酸化珪素粒子 (ヒュームドシリカ) (日本国 日本アエロジル (株) 製、商品名「アエロジル R Y 200」)

(酸化物粒子 - 2)

乾式法で得られた分岐構造を有する酸化珪素粒子 (ヒュームドシリカ) (表面処理されていないもの) (日本国 日本アエロジル (株) 製、商品名「アエロジル 200」)

(酸化物粒子 - 3)

乾式法で得られ、オクチルシランで表面処理された分岐構造を有する酸化チタン粒子 (日本国 日本アエロジル (株) 製、商品名「アエロジル T 805」)

3. 成分 (C) : 有機スルホン酸アルカリ金属塩

( $C_4F_9SO_3K$ )

パーフルオロブタンスルホン酸カリウム (日本国大日本インキ工業 (株) 製 商品名「メガファック F 114」)

4. 成分 (D) : フルオロポリマー